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Results of the California Healthy Homes Indoor Air Quality Study of 2011-2013: Impact of Natural Gas Appliances on Air Pollutant Concentrations

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ABSTRACT

This study was conducted to assess the current impact of natural gas appliances on air quality in California homes. Data were collected via telephone interviews and measurements inside and outside of 352 homes. Passive samplers measured time-resolved CO and time-integrated NO_X , NO_2 , formaldehyde, and acetaldehyde over \sim 6d periods in November 2011 – April 2012 and October 2012 – March 2013. The fraction of indoor NO_X and NO_2 attributable to indoor sources was estimated. NO_X , NO_2 and highest 1-h CO were higher in homes that cooked with gas and increased with amount of gas cooking. NO_X and NO_2 were higher in homes with cooktop pilot burners, relative to gas cooking without pilots. Homes with a pilot burner on a floor or wall furnace had higher kitchen and bedroom NO_X and NO_2 compared to homes without a furnace pilot. When scaled to account for varying home size and mixing volume, indoor-attributed bedroom and kitchen NO_X and kitchen NO_2 were not higher in homes with wall or floor furnace pilot burners, though bedroom NO_2 was higher. In homes that cooked 4 h or more with gas, self-reported use of kitchen exhaust was associated with lower NO_X , NO_2 and highest 1-h CO. Gas appliances were not associated with higher concentrations of formaldehyde or acetaldehyde.

Keywords: Carbon monoxide; Cooking; Formaldehyde; Natural gas appliances; Nitrogen dioxide; Kitchen Ventilation

PRACTICAL IMPLICATIONS

The findings (1) that use of natural gas cooking burners substantially increases the risk of elevated CO, and (2) that gas cooking and the presence of pilot burners on cooking and heating appliances within the living space are associated with elevated NO_X and NO_2 are consistent with prior studies and demonstrate that there is still a need to address these indoor air quality challenges in California (and likely other U.S.) homes. Smaller homes are more impacted by pollutant emissions from unvented cooking and pilot burners. Study results suggest that IAQ benefits would result from accelerating replacement of existing appliances with pilot burners and ensuring that suitable exhaust hoods or kitchen fans are installed and routinely used. California's state building code currently requires kitchen exhaust ventilation for all new homes, but codes in most U.S. states do not. And millions of existing homes lack any kitchen exhaust ventilation. Results indicate that venting appliances do not frequently release pollutants into the home in the amounts necessary to increase time-averaged concentrations.

INTRODUCTION

Residential natural gas appliances can produce pollutants including carbon monoxide (CO), nitrogen dioxide (NO₂), formaldehyde and ultrafine particles (UFP) (Afshari et al., 2005; Brown et al., 2004; Dennekamp et al., 2001; Moschandreas et al., 1986; Singer et al., 2010; Traynor et al., 1996; Traynor et al., 1985). When the exhaust from a gas appliance enters the living space, indoor air quality (IAQ) can be compromised. Many gas appliances, including water heaters and furnaces, are designed to vent their exhaust directly to the outdoors. If the venting is not operating correctly – e.g., because it is broken or not designed and installed correctly, or when depressurization in the indoor space exceeds the draft capacity of the appliance – combustion products including pollutants spill into the indoor space. Combustion products of cooking appliances and "vent-free" (unvented) heating appliances are released

indoors by design. Venting range hoods (extractor fans) and other kitchen exhaust fans are intended to remove some of the pollutants emitted by cooking burners before they mix throughout the home (Delp and Singer, 2012; Singer et al., 2012). However, surveys suggest that regular use of kitchen ventilation during cooking is infrequent, even when it is available (Klug et al., 2011; Mullen et al., 2013a; Piazza et al., 2007).

Numerous studies have found that homes with gas cooking burners and/or gas appliances with pilot burners tend to have indoor concentrations of combustion-related pollutants that are higher than similar homes without gas appliances, and that sometimes exceed U.S. national and California state ambient air quality standards (AAQS) (Garrett et al., 1999; Ryan et al., 1988; Schwab et al., 1994; Spengler et al., 1994; Spengler et al., 1983; Wilson et al., 1986; Wilson et al., 1993). A recent simulation study estimated that among Southern California homes that cook at least once per week with natural gas and do not regularly use a venting range hood, more than half have 1-h NO2 concentrations exceeding 100 ppb and roughly 5% have short-term CO concentrations that exceed the concentration thresholds of acute ambient standards on a weekly basis in winter (Logue et al., 2014). Homes that use unvented gas heaters and fireplaces can have particularly high concentrations of combustion pollutants, often exceeding AAQS thresholds (Dutton et al., 2001; Francisco et al., 2010; Ryan et al., 1989). In homes with gas appliances, smaller home size and the presence of floor and wall furnaces have been associated with higher combustion pollutant levels (Wilson et al., 1986).

There is a large literature showing associations between exposure to pollutants generated by gas appliances and adverse health impacts, with many of the studies focusing on nitrogen dioxide (Belanger et al., 2006; Franklin et al., 1999; Garrett et al., 1998; Hansel et al., 2008; Morales et al., 2009; Neas et al., 1991; Nitschke et al., 1999; Pilotto et al., 1997; van Strien et al., 2004). The most recent EPA assessment for carbon monoxide concluded that "a causal relationship is likely to exist between relevant short-term exposures to CO and cardiovascular morbidity, whereas the available evidence is inadequate to conclude that a causal relationship exists between relevant long-term exposures to CO and cardiovascular morbidity" (US EPA, 2010). Formaldehyde is a known human carcinogen (International Agency for Research on Cancer, 2006) and exposures at levels that occur in homes have been linked to respiratory pathology (Franklin et al., 2000; Roda et al., 2011). A recent study found higher lung function and lower odds of asthma, wheeze, and bronchitis among children whose parents reported using kitchen ventilation when cooking with gas compared to children living in homes in which kitchen ventilation was not used with gas stoves (Kile et al., 2014).

More than two decades have elapsed since the last large-scale studies that focused on the impacts of natural gas appliances on IAQ in California homes (Spengler et al., 1994; Wilson et al., 1993). During this time, there have been many changes to the population of homes and gas appliances. Burner and appliance designs have advanced and attention to IAQ by appliance manufacturers, utilities and the home renovation industry may have reduced the frequency of improper appliance operation or venting, leading to fewer homes with elevated concentrations. Air sealing retrofits and the construction of new homes with airtight envelopes for energy efficiency should translate to lower outdoor air exchange rates during winter conditions when windows are closed; this could produce higher concentrations of any pollutants that are released into the home.

The California Healthy Homes Indoor Air Quality Study of 2011-2013 was designed to

investigate the extent to which gas appliances still negatively impact IAQ in California homes. The study targeted homes with one or more gas appliances that could be a source of indoor air pollutant emissions, including gas cooking burners and venting appliances contained in the living space. There was oversampling of homes with previously identified risk factors, such as smaller floor area, frequent cooking with gas burners, presence of a wall or floor furnace, and lower household income, as these households can less frequently update or upgrade appliances. This paper presents analyses examining the impact of the types of appliances present in the home, the presence of pilot burners, the frequency of cooking with gas or electric burners and the use of kitchen exhaust during cooking.

MATERIALS AND METHODS

The core data collection methods of the study entailed monitoring inside and outside of homes using passive measurement devices while also conducting telephone interviews with participants to collect information about the homes. Mullen et al. (2013a) provides a thorough description of experimental methods, participant communication materials and all interview questions. The sections below provide summary descriptions. The study protocols were approved by LBNL's Institutional Review Board.

Participant recruitment

The study was publicized by direct outreach to organizations associated with ethnically, economically and geographically diverse sub-populations in California. Recruitment efforts in the first year focused on the northern coastal region of California. The second year focused on the southern and inland regions of the state. Organization representatives were asked to pass along information about the study to their constituents. Interested individuals were directed to a project web site and telephone number to obtain more information and complete a screening survey. The web site noted the incentive of \$75 and a report about the air quality in the participant's home to be provided at the completion of participation. The screening survey asked questions about the building and appliances, household demographics, and activities related to appliance use. Responses were used to calculate a risk score for IAQ hazards from gas appliances based on the algorithm described in Supporting Information Table S1. The following factors were considered: frequency of use of gas cooking burners; which gas appliances were inside the living space or connected spaces and whether they were vented; size of the home; year the home was built (recognizing that newer homes are generally tighter with less infiltration air exchange); household income; and whether the home had been weatherized to increase airtightness. Twenty-four homes constructed or retrofitted for low energy use were included as part of a supplemental study of IAQ in high performance homes (Less, 2012; Less et al., 2014). There was intentional sampling of some homes without gas appliances to serve as controls (n=38). Homes were selected for sampling in geographic clusters. When a home was identified as desirable for inclusion, the individual who submitted the screening survey was contacted by telephone for consent and scheduling. The content of the web site, outreach materials and screening survey is provided in Mullen et al. (2013a).

Data collection instruments and methods

Measurement devices were deployed in homes to determine pollutant concentrations, temperature (T) and relative humidity (RH) in two indoor locations and at an outdoor site nearby to each residence. Furnace and water heater operation were also monitored. A

structured interview was conducted by telephone before monitoring to collect more detailed information about the building, appliances, household demographics and general activities. A post-monitoring structured interview collected data about activities during the monitoring period and about general practices relevant to gas appliance impacts on IAQ. Some questions were not asked until after the monitoring period, so as to avoid affecting occupants' behaviors and attitudes related to their gas appliances.

Measurements were conducted using a package of passive samplers and monitors that were mailed to 323 participant homes and delivered by researchers to 29 homes. Participants receiving the package by mail set up the samplers using written and pictorial instructions provided with the package. A researcher contacted each participant by telephone to check if the materials were clear and to help resolve any difficulties. Monitoring was planned to occur in each home for six days. The standard schedule was for the package to be sent on Monday morning to arrive at the home by Tuesday afternoon. The request was for the samplers to be set up within 24 h of receipt and then repackaged and mailed back the following Tuesday. Participants were asked to package samplers in pre-addressed return shipping envelopes on Monday night or Tuesday morning. In 29 homes, equipment was deployed and retrieved by a researcher who visited the homes. Sampling was conducted in two phases from late November 2011 to mid-April 2012 and from late October 2012 to mid-March 2013. During those periods, 5 to 14 homes were sampled per week during most weeks. Sampling did not occur during the weeks in which the Thanksgiving, Christmas and New Years holidays were observed in the United States.

The monitoring package included samplers and instruments listed in Table 1. Pollutant concentrations, T, and RH were measured in the kitchen and a bedroom (child's bedroom, if available) of each home, and outside of selected homes to define outdoor concentrations for a cluster of similarly located homes. NO₂, NO_x, volatile aldehydes and CO were measured in the kitchen, and all pollutants other than CO were measured in a bedroom. Volatile aldehydes were measured with a sampler that is typically used for active sampling, based on passive uptake rates determined for 5-10 d deployment periods (Mullen et al., 2013b). NO_x and NO₂ were measured using Ogawa passive sampling equipment (Singer et al., 2004), with NO calculated as the difference between the NO_x and NO₂ results.

A thermocouple placed on the water heater and a thermistor placed on a heating supply register monitored the operation of these appliances. Temperature, RH, CO, and appliance monitors all had on-board data loggers. Participants were asked to take photos of the samplers deployed in the homes and to send the photos via email or text message to the study director to ensure proper placement. Most sent relevant photos. Roughly half the homes received either a duplicate sampler that was to be placed in the bedroom or a field blank. Participants were called as a reminder the night before they were expected to return the package.

The post-monitoring telephone interview collected data on activities in the home during the sampling period, including frequency of appliance use, occupancy patterns and other potential pollutant sources inside and outside of the home. The interview included questions that might have affected resident behavior if asked prior to the sampling periods, e.g., about the frequency of kitchen exhaust fan use, reasons why the kitchen exhaust fan was not used, and the condition of the stovetop and oven (flame quality, operational problems etc.). The post-monitoring interview was the last task for participants to complete.

Data analysis

Passive samples were analysed using methods described in (Mullen et al., 2013a). Passive samplers that were returned unsealed were flagged as invalid. Photos and analytical results were reviewed to identify obvious errors such as a samplers being deployed with caps in place or switching of samples and blanks. Data from the CO, T, RH and appliance monitoring data loggers were downloaded and compiled into a database and analysed to calculate mean, as well as the highest 1-h and 8-h averages for the sampling period in each home.

The potential for depositional losses of NO and NO_2 inside the two designs of outdoor sampling enclosures was evaluated in six side-by-side deployments with the open samplers used in Singer et al. (2004); details are reported in Mullen et al. (2013a). Adjustment factors of 1.22 and 1.18 were determined for NO_2 sampling in the outdoor enclosures used in the first two weeks and all subsequent weeks, respectively. The data did not show any clear bias in NO measured in the outdoor enclosure, so no adjustments were made for NO. Outdoor NO_X was calculated as the sum of the adjusted NO_2 and the unadjusted NO (Mullen et al., 2013a).

Recognizing that outdoor NO_X and NO₂ concentrations have a major impact on indoor levels, we used concurrently measured outdoor concentrations to estimate the indoor levels that could be attributed to indoor sources. This adjustment was made for NO_X and NO₂ since outdoor concentrations were of similar magnitude to indoor concentrations (Figure 1). Outdoor air contributed a minority of indoor aldehydes (Figure 2); analyses were thus conducted on the directly measured levels of these pollutants in kitchens and bedrooms. The highest 1-h and 8-h CO in kitchens also were analysed as measured since outdoor levels are typically much lower than short-term indoor peaks in homes with a CO source. Homes without outdoor monitoring were assigned the outdoor NO_X and NO₂ concentrations measured at the closest home within the cluster or the closest ambient monitoring station, when either the cluster sample was not available or the central monitoring site was deemed more representative based on land-use. Indoor concentrations attributed to indoor sources were calculated as follows. For NO (NO_X-NO₂), outdoor levels were subtracted from those measured indoors. For NO₂, we multiplied the assigned outdoor value by an infiltration factor F=0.4 to obtain an estimate of the indoor NO₂ that can be attributed to outdoor sources. This value is obtained as the air exchange rate (λ) – accounting for entry from outdoors to indoors - divided by the sum of the air exchange rate and indoor deposition rate $(\lambda + k_d)$ - which is the rate at which NO₂ is removed from indoors. The value of 0.4 was estimated based on consideration of published data on air exchange rates in California homes (Wilson et al., 1993; Wilson et al., 1996; Yamamoto et al., 2010) and reported NO₂ indoor deposition rates (Noris et al., 2013; Spicer et al., 1989; Spicer et al., 1993; Wilson et al., 1986; Yang et al., 2004). Indoor NO_X attributed to entry from outdoors was calculated as the sum of NO and NO₂ from outdoors. Figure 3 and Supporting Information Table S4 show that after the estimated outdoor contribution is subtracted, the median bedroom NO₂ and NO_X in allelectric homes were both close to zero, as would be expected for homes with no indoor sources. s

The impacts of gas appliances on IAQ was explored by comparing distributions of calculated pollutant concentrations noted above grouped by the following characteristics: (a) the type(s) of gas appliance(s) inside the living space, (b) cooking burner fuel type and which appliances, if any, had pilot burners, (c) cooking burner fuel type and frequency of use and

(d) use frequency of kitchen exhaust ventilation in homes that reported cooking for 4 or more hours during the monitoring period. Analyses were conducted using the measured, time-integrated concentrations of aldehydes, the highest 1-h and 8-h CO, and the estimated indoor source-attributed concentrations of NO_X and NO_2 .

Recognizing that the impact of emissions from a combustion appliance or pilot burner will scale inversely with the dilution volume, we scaled the indoor-attributed concentrations of NO_X and NO_2 and measured CO to a common home size of 130 m² (1400 ft²). This scaling was done after the first series of bivariate analyses revealed that homes with gas cooking appliances and with pilot burners had significantly higher concentrations of these pollutants than homes without gas cooking. This analysis was designed to assess if any between-group differences in un-scaled concentrations were caused by differences in homes sizes.

RESULTS AND DISCUSSION

Demographics of Sample

Data were collected from 352 homes, including the high performance home sub-sample (Less, 2012). The overall sample mostly comprised homes with gas appliances in the living space and homes that used gas cooking appliances: 90% of study homes had at least one gas appliance and 82% had gas cooking burners. A gas cooktop was used more than 7 times during the sampling period in 53% of study homes and 26% of study homes used a gas cooktop more than 14 times (all by self-report). Participants reported that they either did not have a kitchen exhaust fan or that they rarely or never used it in 64% of homes.

The sample included many older appliances, as reported in Mullen et al. (2013a). Table 19 of that report indicates that 24% (40/165) of central furnaces and 65% (35/54) of wall and floor furnaces with estimated ages were more than 15 years old. Table 25 of the report indicates that 20 of 150 water heaters (13%) with age estimates were more than 15 years old. And Table 38 of the report indicates that 20% (62/310) of cooktops with age estimates were more than 15 years old.

The demographics of the mail-out sample population are presented and discussed by Mullen et al. (2013a) and summarized in Table S2 of Supporting Information. The study sample had a similar breakdown of renters and homeowners (46/54%) compared to California overall (43/57%) (RASS, 2009). The sample had more homes with floor areas under 93 m², fewer homes larger than 186 m² and similar percentages of 93-186 m² homes compared to the California stock. The study sample was under-represented in the lowest household income brackets (<\$50,000 per year), with 19% in the sample compared to 44% for the state. Although we could not find directly comparable statewide data, it seems likely that the educational attainment of the study sample was skewed relative to the general population. The racial distribution of the sample was reasonably similar to that of the California population, allowing for uncertainty related to the US Census not tracking "Hispanic" as a race and considering that census data is tabulated per individual whereas statistics on the study population are tabulated per household. Relative to California, there were fewer households in the study containing children or seniors.

Quality Assurance Results

The available evidence – including survey completion and sampler return rates, submitted photographs of sampler deployment locations, inspections of returned sampler packages and

results of quality assurance replicates and blanks – indicates that most participants followed the instructions to deploy samplers as intended (Mullen et al., 2013a). In only one instance did a participant report that a sampling package mailed from LBNL did not arrive. Sampler packages were mailed back by all participants who received them. In seven cases, data were lost from all passive samplers sent to a home, either because participants returned the package with delays of more than a month, or participants did not seal time-integrated samplers in the provided airtight bags before mailing. Two additional homes had invalid NO_X and NO₂ data because of an error in sampler preparation before shipment to one home and improper sealing of the samplers from the other home. The mean relative deviations for all pairs of duplicate samplers were 3% for NO_X, 7% for NO₂, 5% for formaldehyde and 5% for acetaldehyde. The percent of field blanks with concentrations above the analytical LOQ were 8%, 5%, 16% and 45% for NO_X, NO₂, formaldehyde and acetaldehyde. Field blanks had mean concentrations of 0.37 ppb NO_x, 0.25 ppb NO₂, 0.6 ppb formaldehyde and 1.7 ppb acetaldehyde for an assumed 6-day deployment period. Reported measured concentrations were not adjusted for the values measured on field blanks. Additional quality assurance results and participant compliance notes are presented and discussed in Mullen et al. (2013a).

Measured Pollutant Levels in Kitchen, Bedroom and Outdoors

Summary statistics for all measured pollutants and pairwise correlations are provided in Supporting Information Tables S3 and S4.

The time-integrated concentrations of NO_X and NO₂ measured indoors and measured or assigned outdoors at each home are presented in Figure 1. Each series (outdoor, bedroom and kitchen) follows a lognormal distribution, as shown in Supporting Information Figure S1. Outdoor NO₂ concentrations were higher than the 30 ppb threshold of California's annual average ambient air quality standard (CAAQS) for 9% of study homes. Measured NO₂ exceeded 30 ppb in about 24% of kitchens and 12% of bedrooms, and indoor-attributed NO₂ was above 30 ppb in about 14% of kitchens and 6% of bedrooms. These statistics result from monitoring over periods of only about 6 days in each home and over-sampling of homes with potential indoor sources of NO₂. Figure 1 and Supporting Information Table S4 show that concentrations of NO_X ($r^2=0.90$) and NO_2 ($r^2=0.86$) were highly correlated between kitchens and bedrooms. Many homes had higher NO_X in bedrooms and kitchens than outdoors, indicating indoor source(s). In the absence of indoor sources, indoor NO₂ should be substantially lower than outdoor NO₂ owing to indoor deposition. The homes with the lowest values of bedroom NO₂ had indoor concentrations that were on the order of half of outdoor levels. At higher bedroom NO₂ concentrations, the ratio of indoor to outdoor NO₂ generally was higher. For NO₂, there was a clear trend of higher concentrations in the kitchen than in the bedroom: arithmetic (AM) and geometric (GM) mean levels of NO₂ in kitchens (23.2 and 16.9 ppb) were 31% and 26% higher than NO₂ in bedrooms (17.7 and 13.4 ppb). Kitchen NO_X was also higher than bedroom NO_X, with AM and GM ratios of 13% and 14%.

Broadly, NO₂ concentrations measured in the Healthy Homes study of 2011-2013 were lower than those reported for California homes in large studies conducted in the 1980s and early 1990s (Spengler et al., 1994; Wilson et al., 1986; Wilson et al., 1993), with decreases in outdoor pollutant levels accounting for much or all of the difference. Detailed comparisons for subgroups of homes divided by appliance type are provided in the Supporting Information.

Figure S2 in Supporting Information shows that the highest 1-h and highest 8-h CO levels were log-normally distributed across homes that had CO exceed the instrument quantitation

limit of 0.5 ppm. Of the 316 homes with CO data in the current study, roughly 5% had short term concentrations exceed California ambient air quality standards of 20 ppm over 1 h or 9 ppm over 8 h. Arithmetic and geometric mean values of highest 1-h CO were 6.4 and 3.8 ppm in the current study. These values are similar to those measured or simulated in other studies of California homes, as described in the Supporting Information.

The time-integrated concentrations of formaldehyde and acetaldehyde measured indoors and measured or assigned outdoors at each home are presented in Figure 2. Roughly 95% of homes had indoor formaldehyde levels above the Cal-EPA Chronic Reference Exposure Level (CREL) of 7.3 ppb. Indoor aldehyde concentrations were higher than outdoor concentrations in almost all homes with data for both locations. Concentrations of each pollutant measured in bedrooms and kitchens of the same homes were somewhat correlated with r^2 =0.52 for formaldehyde and r^2 =0.76 for acetaldehyde (Table S4). Formaldehyde and acetaldehyde were not highly correlated with each other, with r^2 =0.34 and r^2 =0.36 for measurements in kitchens and bedrooms. Figure S3 illustrates the lognormal distributions of formaldehyde and acetaldehyde concentrations measured in kitchens, bedrooms and outdoors. As a group, outdoor acetaldehyde concentrations were indistinguishable from field blanks. Comparisons to aldehyde concentrations reported in other California studies are provided in the Supporting Information.

Concentrations of NO_X and NO_2 were not highly correlated with CO or either aldehyde; and the aldehydes were not highly correlated with CO (Table S4).

Impact of Appliance Types on Indoor Pollutant Levels

Figure 3 presents summary statistics for highest 1-h CO in the kitchen, as well as indoor-attributed NO_2 and NO_X and formaldehyde and acetaldehyde in the bedroom, grouped by the type(s) of appliances inside the home. P-values in the figure represent the likelihood that other groups' distributions are drawn from the same distribution as the "All Electric" group, based on the Kruskal-Wallis test. Table S5 in the Supporting Information presents additional results for this analysis. In comparison to homes without gas appliances, there was a large and statistically robust increase in indoor-attributed concentrations of bedroom and kitchen NO_X and NO_2 and highest kitchen 1-h CO for homes that used gas cooking burners, whether or not there were also venting gas appliances in the home. Indoor-attributed NO_X and NO_2 concentrations were higher in kitchens than in bedrooms for the two groups with gas cooking appliances. Table S5 shows that in comparison to homes with gas cooking but no venting appliances, homes with both cooking and venting appliances had significantly higher indoor-attributed NO_X and NO_2 . Highest 1-h kitchen CO was not different between these groups.

Some of the differences in NO_X and NO_2 between the last two groups of Figure 3 and Table S5 may result from differences in home volumetric dilution rates. The outdoor air dilution rate (e.g. in units of m^{-3} h^{-1}) is the product of the residence air volume and the air exchange rate. An air pollutant source of fixed size, such as a cooking burner, will have less dilution in a smaller home compared to a larger home with the same outdoor air exchange rate. When concentrations were scaled to home size (by floor area), the difference in NO_X and NO_2 between the last groups disappeared (see last 4 rows of Table S5). This suggests that much / all of the difference between those groups may result from cooking burner pollutant emissions occurring in smaller spaces with less outdoor air dilution.

There were no statistically robust differences in formaldehyde or acetaldehyde levels

associated with gas appliances (Table S5).

Impact of Pilot Burners on Indoor Pollutant Levels

Figure 4 and Table S6 present summary statistics for the same pollutants displayed in Figure 3 and Table S5, this time grouped by cooktop fuel type and the presence of pilot burners on cooktops or furnaces located inside the home. The homes were divided into five groups: (1) electric cooktop, no furnace pilots; (2) gas cooktop without pilot, no furnace pilots; (3) gas cooktop with pilot, no furnace pilots; (4) gas cooktop without pilot, furnace(s) with pilot(s); (5) gas cooktop, furnace(s) with pilot(s). The fourth group includes seven homes with floor furnaces and 41 homes with wall furnaces, four of which did not have valid NO₂ and NO_x data. The fifth group includes 3 homes with floor furnaces and 24 with wall furnaces, one of which did not have valid NO₂ and NO_x data. Each group of homes was compared to homes that had gas cooking but no pilot burners, using the Kruskal-Wallis test, with p-values shown in the figure. We also compared the third and fifth groups to further explore whether homes with furnace pilots have higher pollutant concentrations than those without.

All three groups with any pilot burner had indoor-attributed NO_X and NO_2 in bedrooms (Figure 4) and kitchens (Table S6) that were significantly higher than homes with gas cooktops but no pilots (Group 2). Higher concentrations in Group 4 compared to Group 2 (with p-values of 0.02 to <0.01) suggests that furnace pilots significantly increase NO_X and NO_2 throughout the home. The impact of furnace pilot burners is further indicated by higher concentrations in homes with both cooking and furnace pilots (Group 5) compared to homes with gas cooking pilots only (Group 3); Table S6 shows that differences in indoor-attributed NO_X and NO_2 between these groups are significant with p-values of <0.01 to 0.03 for three of the parameters and p=0.09 for kitchen NO_X .

The three groups of homes with pilot burners also appear to have had higher values of the highest 1-h kitchen CO compared to the gas cooktop homes with no pilots, with p-values of 0.01-0.09. Bedroom formaldehyde was *lower* in the last two groups with p-values of 0.06 and 0.09.

Much of the apparent impact of furnace pilots appears attributable to these appliances being present in smaller homes, which may have lower volumetric dilution rates as noted earlier. The last 4 rows of Table S6 show that differences in NO_X and NO_2 between homes with gas cooktops and only furnace pilots (Group 4) and gas cooktops with no pilots (Group 2) largely disappear when indoor-attributed concentrations are adjusted by the size (floor area) of the home. The effect of furnace pilots in homes that also have cooktop pilots – comparing Groups 3 and 5 – persists for bedroom NO_2 (p=0.03), but not for bedroom NO_X or kitchen NO_X and NO_2 , when adjusting for floor area.

Impact of Cooking Burner Use on Indoor Pollutant Levels

Figure 5 and Table S7 present summary pollutant statistics for homes grouped according to cooking appliance fuel and cooking time during the monitoring period. Cooking time was estimated as the sum of self-reported cooking activity by meal. Highest 1-h kitchen CO and indoor-attributed NO_X and NO₂ measured in both kitchens and bedrooms increased with more gas cooking but not with more electric cooking. This trend was seen with and without scaling for floor area (Table S7). Formaldehyde and acetaldehyde in homes that cooked more frequently with gas appliances were statistically indistinguishable from those that cooked less

frequently with gas or cooked with electric appliances at any frequency (Table S7). These results add to the weight of evidence that natural gas cooking burners are substantial and statistically significant sources of CO, NO_X and NO₂ in many homes.

Impact of Kitchen Exhaust Ventilation on Indoor Pollutant Levels

The final bivariate analysis investigated the impact of using kitchen exhaust fans when cooking. For this analysis, homes that reported cooking with gas for more than 4 h total during the week were grouped according to self-reported frequency of kitchen exhaust fan use. The Kruskal-Wallis test was used to compare homes in which kitchen exhaust was used some times or most times when cooking with gas against homes that cooked with gas but either never used or did not have a kitchen exhaust fan. Figure 6 presents summary statistics for highest 1-h kitchen CO and indoor-attributed kitchen NO₂ and NO_X; additional results are presented in Supporting Information Table S8. Measured aldehydes were not included in the analysis, since the prior analyses showed they were not significantly influenced by gas cooking in the homes. The results suggest that even occasional use of a kitchen exhaust fan reduces peak CO in the kitchen and time-integrated NO₂ and NO_X throughout the home. The effect broadly persists but at lower significance levels (higher p-values) when indoorattributed concentrations are adjusted for home floor area (Table S8). The lack of a clear progression from infrequent to frequent use could be related to how decisions are made about exhaust fan use. For example, occasional use may occur during the most intensive cooking events, having a disproportionate effect on both peak and time-integrated concentrations in the home. The very wide range in pollutant removal effectiveness for range hoods installed in existing California homes (Singer et al., 2012) might also have obscured the expected relation between pollutant concentrations and frequency of range hood usage, such that consistent usage in some homes may have very low efficacy.

These results provide empirical evidence that regular use of a kitchen exhaust fan when cooking with gas burners helps reduce concentrations of combustion pollutants in the kitchen. The effectiveness of range hoods in these homes presumably was reduced by the fact that 35% of participants (among those having fans) reported using it on medium or low speed, and 70% of participants reported cooking primarily on front burners. Research on range hood effectiveness indicates that the effectiveness is substantially lower when the hoods are operated at lower speeds and when cooking occurs on the front burners (Delp and Singer, 2012; Lunden et al., 2014; Singer et al., 2012).

CONCLUSIONS

Pollutant measurements over multiple day monitoring periods in 352 California homes demonstrate that associations still exist between the presence and use of some gas appliances and elevated concentrations of CO, NO_X and NO_2 . The largest impacts were associated with use of gas cooking appliances. More cooking led to higher concentrations in homes with gas cooking appliances but not in homes with electric cooking. In homes with gas cooking, the presence of additional appliances with venting was associated with higher concentrations of indoor-attributed NO_X and NO_2 . However, when indoor-attributed concentrations were scaled to home size – to account for pollutants from cooking burners possibly reaching higher concentrations in smaller homes owing to less overall dilution – the effect of vented appliances on NO_X and NO_2 disappeared. Cooktop and furnace pilot burners were each associated with higher concentrations of time-integrated, indoor-attributed NO_X and NO_2 and highest 1-h CO when not scaled for home size. When pollutant concentrations were scaled to

a common home size, the impacts of furnace pilot burners largely disappeared. Formaldehyde and acetaldehyde concentrations were not significantly impacted by any of the gas appliances examined in this study. Homes that cooked frequently with gas burners and reported using kitchen exhaust ventilation had lower concentrations of highest 1-h CO and time-integrated NO_X and NO_2 compared to homes that never use kitchen exhaust ventilation when cooking with gas burners.

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Table 1. Summary of pollutant and environmental monitoring instruments used in study.

| Parameter | Manufacturer, model | Data resolution | Location of deployment |
|--|--|-------------------------------|--|
| Formaldehyde, Acetaldehyde ^a | Waters, Sep-Pak XPoSure DNPH Cartridges | Integrated over sample period | Bedroom, kitchen, outdoor ^a |
| NO _X , NO ₂ ^a | Ogawa NO _X /NO ₂ sampler | Integrated over sample period | Bedroom, kitchen, outdoor ^a |
| CO (ambient) | Lascar, USB-EL-CO300 | 1 minute | Kitchen |
| T, RH (indoors) | HOBO, U10 | 1 minute | Bedroom, kitchen |
| Furnace operation (by T) | HOBO, U10 | 1 minute | Furnace supply register |
| Water heater operation (T) | HOBO, U12-014 | 1 minute | Water heater exhaust flue |
| Water heater spillage (T) | HOBO, U12-014 | 1 minute | Adjacent to draft hood |
| T, RH (outdoors) ^a | HOBO, U23 Pro v.2 | 1 minute | Outdoors |

^a Outdoor sampling occurred at a subset of homes.

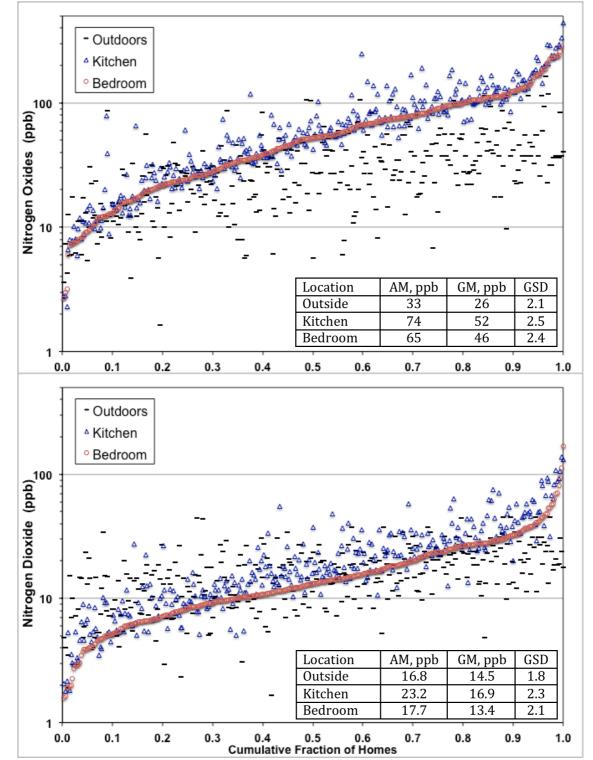


Figure 1. NO_x and NO₂ measured in kitchen and bedroom, and measured or assigned

outdoor concentrations, ordered by concentrations in bedroom.

Data displayed for 343 homes with bedroom measurements; results for each home aligned vertically. Outdoor concentrations were measured in this study or taken from a nearby regulatory air monitoring station. Figure S1 of supporting information shows that data at each location follow lognormal distributions. Tables present arithmetic means, geometric means, and geometric standard deviations.

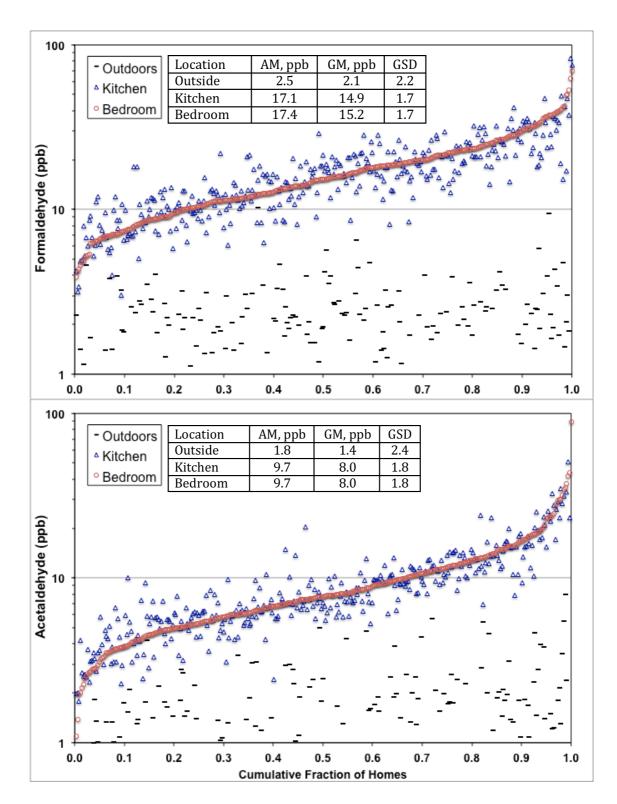


Figure 2. Formaldehyde and acetaldehyde measured in kitchens, bedrooms, and outdoors of study homes, ordered by bedroom concentrations.

Data displayed for 344 homes with bedroom measurements; results for each home aligned vertically. Outdoor concentrations were measured only at a subset of homes. Figure S2 of supporting information shows that data at each location follow lognormal distributions. Tables present arithmetic means, geometric means, and geometric standard deviations. As a group, outdoor acetaldehyde concentrations were indistinguishable from field blanks.

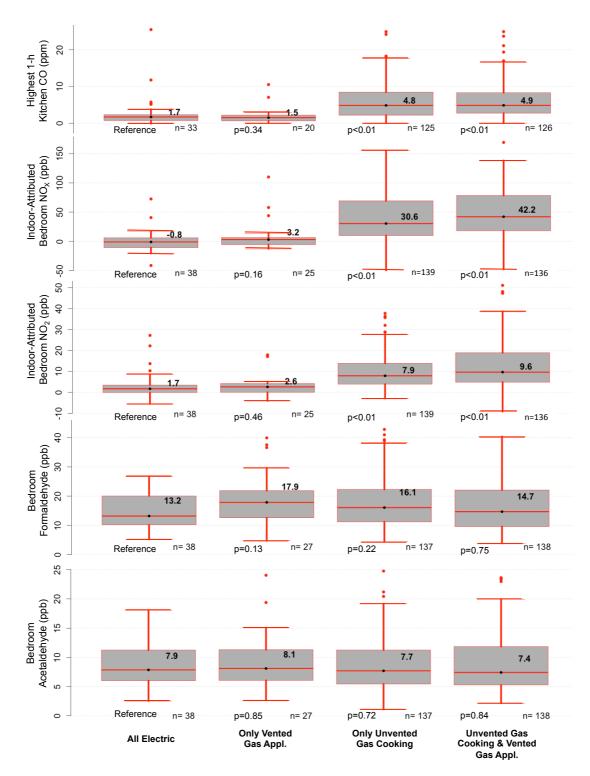


Figure 3. Indoor pollutant concentrations by type(s) of appliances inside home. Highest 1-h CO from kitchen and indoor-attributed NO_X and NO₂ from bedroom measurements. Formaldehyde and acetaldehyde from bedroom measurements. Indoor-attributed concentrations calculated by subtracting estimated outdoor contribution from the indoor measured value. See text for additional details. Boxes show inter-quartile range (IQR). Whiskers span to 1.5 IQR. Filled circles show all data >1.5 IQR. P-values indicate likelihood that data from other groups are drawn from same distribution as the "All Electric" group, based on the Kruskal-Wallis test.

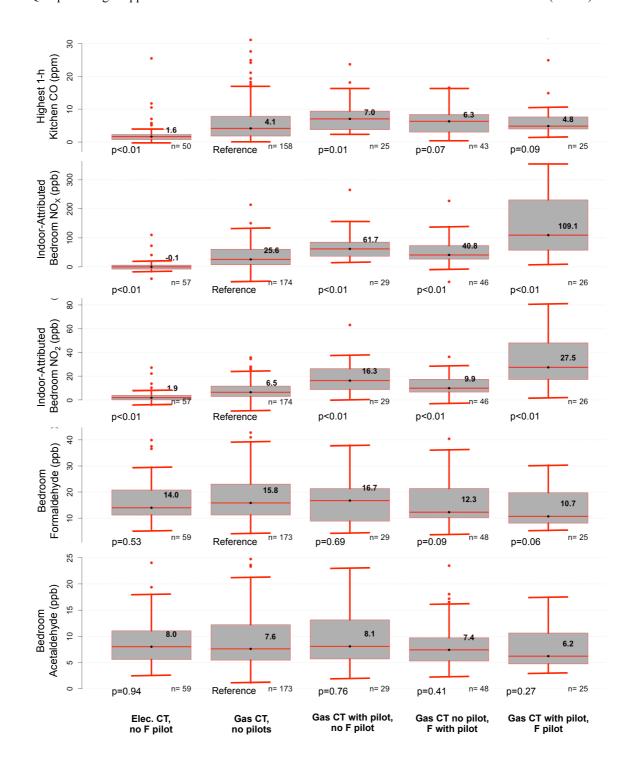


Figure 4. Indoor pollutant concentrations by cooktop (CT) fuel and presence of pilot burners on cooktop or furnace (F).

Refer to Figure 3 caption and text for descriptions of calculations for indoor source attribution and definitions of boxes and whiskers. P-values indicate likelihood that data from other groups are drawn from the same distribution as the "Gas CT, no pilots" group, based on the Kruskal-Wallis test.

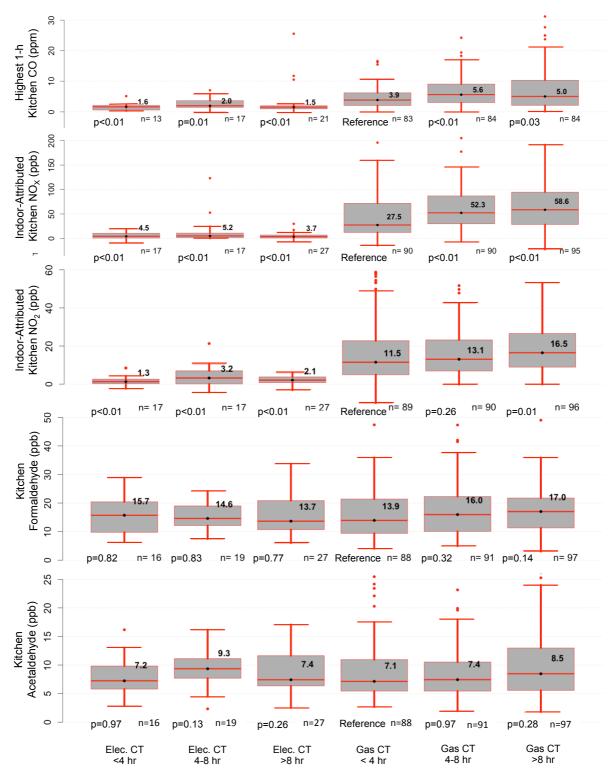


Figure 5. Indoor pollutant concentrations by cooktop (CT) fuel and respondent-reported total cooking time during monitoring period.

Cooking time from daily log. Refer to Figure 3 caption and text for descriptions of calculations for indoor source attribution and definitions of boxes and whiskers. P-values indicate likelihood that data from other groups are drawn from same distribution as the "Gas CT, <4 h" group, based on the Kruskal-Wallis test.

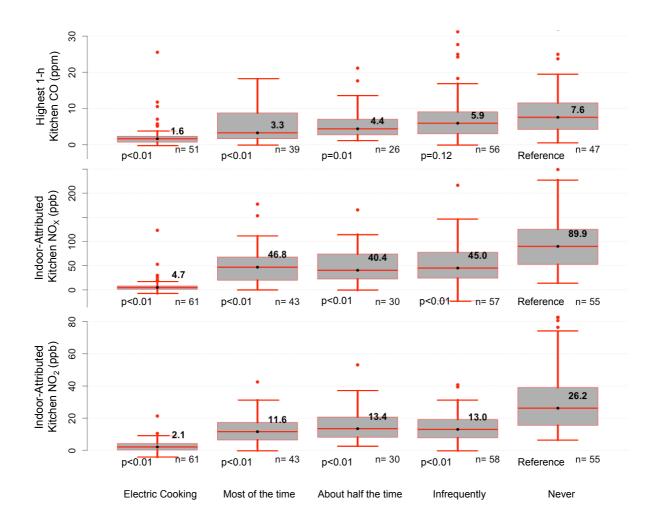


Figure 6. Indoor pollutant concentrations by kitchen exhaust fan use during cooking in homes with gas cooktops and >4 h cooking.

Cooking time from daily log. Exhaust fan use reported by respondent. Refer to Figure 3 caption and text for descriptions of calculations for indoor source attribution and definitions of boxes and whiskers. P-values indicate likelihood that data from other groups are drawn from same distribution as the "Gas CT, <4 h" group, based on the Kruskal-Wallis test.

Supporting Information

Table S1. Algorithm for calculating a "hazard score" used for recruiting homes that have risk factors for indoor pollutant impacts from gas appliances.

The scores used data provided by potential participants via an online screening survey.

| Points for gas cooking a | Points for gas cooking appliances based on amount of use | | | | | | | | |
|--|--|-----------------|---------------|----------|--|--|--|--|--|
| | <1x/wk | 1-3x / wk | 4-7x / wk | >7x/wk | | | | | |
| Cooktop | 1 | 1.5 | 2 | 3 | | | | | |
| Oven | 1 | 1.5 | 2 | 3 | | | | | |
| Points for primary gas | heater (eval | uate per appl | liance). | | | | | | |
| Unvented heater ^a in living space | 3 | | | | | | | | |
| Unvented heater in adjacent space ^b | 1.5 | | | | | | | | |
| Vented gas heater in living space | 1 | | | | | | | | |
| Vented gas heater in adjacent space b | 0.5 | | | | | | | | |
| Points for supplementary | gas heater (e | evaluate per a | ppliance). | | | | | | |
| Unvented heater in living space | 2 | | | | | | | | |
| Unvented heater in adjacent space ^b | 1 | | | | | | | | |
| Vented gas heater in living space | 0.5 | | | | | | | | |
| Vented gas heater in adjacent space b | 0 | | | | | | | | |
| Points for gas storage water heater pe | er number of | f residents (ev | aluate per ap | pliance) | | | | | |
| | 1-2 people | 3-4 people | 5+ people | | | | | | |
| Vented water heater in living space | 0.5 | 1 | 1.5 | | | | | | |
| Vented water heater in adjacent space b | - | 0.5 | 1 | | | | | | |
| Multiplier for oth | er household | characteristi | ics | | | | | | |
| (Sum points for categories below, ad | | tiply by sum o | · | above) | | | | | |
| Year home was built | < 1995 | 1995-2005 | > 2005 | | | | | | |
| | - | 0.1 | 0.2 | | | | | | |
| Size of home (square feet) | < 500 | 500-1000 | 1000-1500 | >1500 | | | | | |
| | 0.3 | 0.2 | 0.1 | - | | | | | |
| Household gross income (\$1000/year) | < 30 | 30-60 | >60 | | | | | | |
| | 0.3 | 0.1 | <u>-</u> | | | | | | |
| Weatherization renovations | No | Yes | | | | | | | |
| | | | | | | | | | |

^a Included use of gas oven for space heating.
^b Adjacent space" includes attic, basement or attached garage.

Table S2. Self-reported race and/or ethnicity, household income, highest education level and number of residents living in households included in this study.

| | # in study | % in study | % in CA a |
|--|------------|------------|------------------|
| Types of appliances present | | | |
| Home rented | 147 | 46% | 43% |
| Home owned | 176 | 54% | 57% |
| Floor Area of home (sq. ft.) | | | |
| <1000 | 110 | 34% | 22% |
| 1000-2000 | 143 | 44% | 46% |
| >2000 | 47 | 15% | 32% |
| Did not answer | 23 | 7% | |
| Number of residents | | | |
| 1 – 2 | 164 | 51% | 55% |
| 3 - 4 | 116 | 36% | |
| 5 or more | 42 | 13% | 45% ^b |
| Did not answer | 1 | <1% | |
| Presence of minors and seniors | | | |
| At least one resident <18 years old | 51 | 16% | 37% |
| At least one resident >64 years old | 20 | 6% | 25% |
| All residents between 18-64 years old | 252 | 78% | 38% |
| Highest education level of ANYONE in household c | | | |
| Less than Bachelors degree | 60 | 19% | NA |
| Bachelors degree | 90 | 28% | NA |
| Graduate degree | 172 | 53% | NA |
| Did not answer | 1 | <1% | |
| Ethnicities represented by residents d | | | |
| Native American | 7 | 1% | 2% |
| Hispanic/ Latino | 36 | 5% | 38% |
| Black, African-American | 45 | 14% | 7% |
| Asian or Pacific Islander | 80 | 30% | 14% |
| White, Caucasian | 219 | 76% | 74% |
| Combined Gross Income | | | |
| Less than \$25,000 | 50 | 6% | 22% |
| \$25,000 - \$49,999 | 47 | 13% | 22% |
| \$50,000 - \$74,999 | 53 | 15% | 17% |
| \$75,000 - \$99,999 | 36 | 14% | 12% |
| \$100,000 - \$150,000 | 67 | 25% | 14% |
| >\$150,000 | 36 | 18% | 13% |
| Prefer not to say | 34 | 6% | |

^a Home floor area data obtained from Residential Appliance Saturation Survey, 2009 (www.energy.ca.gov/appliances/rass/). Remaining data obtained from www.census.gov. ^b Percent of households with 3 or more persons in CA.

^c Educational attainment statistics were not available on a per household basis for the CA population.

^d All race/ethnic categories that partially/fully characterize an individual/household are weighted equally, therefore percentages sum to greater than 100%. However, statistics for the study population are tabulated on a per household basis, whereas CA statistics are tabulated per individual.

Table S3. Summary statistics for measured pollutant concentrations.

| Parameter (integrated over sample | N | AM | GM | GSD | 90 th | 95 th |
|---|-----|-----|-----|-----|--------------------|--------------------|
| period, except where noted) | | | | | %-ile ^b | %-ile ^b |
| Kitchen NO _X | 343 | 73 | 51 | 2.5 | 150 | 192 |
| Bedroom NO _X | 344 | 65 | 47 | 2.4 | 124 | 168 |
| Outdoor NO _X , measured ^c | 180 | 32 | 24 | 2.2 | 61 | 70 |
| Outdoor NO _X , assigned ^d | 345 | 32 | 26 | 2.1 | 57 | 84 |
| Kitchen NO ₂ | 343 | 23 | 17 | 2.3 | 108 | 147 |
| Bedroom NO ₂ | 344 | 18 | 13 | 2.2 | 32 | 41 |
| Outdoor NO ₂ , measured ^c | 180 | 16 | 13 | 1.9 | 29 | 33 |
| Outdoor NO ₂ , assigned ^d | 345 | 17 | 14 | 1.8 | 28 | 32 |
| Kitchen highest 8-h CO | 304 | 3.4 | 2.2 | 2.6 | 6.8 | 10 |
| Kitchen highest 1-h CO | 304 | 6.4 | 3.8 | 2.8 | 13 | 18 |
| Outdoor CO, assigned ^d | 334 | 0.5 | 0.5 | 1.5 | 0.9 | 1.0 |
| Kitchen formaldehyde | 340 | 17 | 15 | 1.7 | 29 | 34 |
| Bedroom formaldehyde | 340 | 17 | 15 | 1.7 | 30 | 36 |
| Outdoor formaldehyde, measured ^c | 179 | 2.4 | 2.0 | 2.1 | 3.4 | 3.9 |
| Kitchen acetaldehyde | 340 | 9.7 | 8.0 | 1.8 | 16 | 23 |
| Bedroom acetaldehyde | 340 | 9.7 | 7.9 | 1.8 | 17 | 23 |
| Outdoor acetaldehyde, measured ^c | 178 | 1.8 | 1.4 | 2.4 | 3.2 | 4.6 |

^a Plots of most of the data distributions are provided in Supporting Information
^b Percentiles from measured or assigned values, not fitted distributions
^c Measured in this study
^d Statistics of values assigned to all homes in the study. Assignments based on measurements conducted in this study and values obtained from compliance monitoring sites, as described in text and Mullen et al. 2013.

Table S4. Coefficient of determination (R²) between pollutants measured at different locations in homes.

 NO_2 and NO_X are estimates of the indoor concentrations resulting from indoor sources ^a. Letters "B" and "K" following pollutant abbreviations are used to indicate measurements made in bedrooms and kitchens, respectively. (FA = formaldehyde, AA = acetaldehyde).

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|--------------------|--------------------|--------------------|--------------------|--------------------|-------------|--------------|---------|-----------|------|------|----------|------|
| | NO ₂ -K | NO ₂ -B | NO _X -K | NO _X -B | NO-K | NO-B | CO-1h | CO-8h | FA-K | FA-B | AA-K | AA-B |
| NO ₂ -K | 1.00 | | | | | | | | | | | |
| NO ₂ -B | 0.74 | 1.00 | | | | | | | | | | |
| NO _X -K | 0.79 | 0.61 | 1.00 | | | | | | | | | |
| NO _X -B | 0.61 | 0.74 | 0.83 | 1.00 | | | | | | | | |
| NO-K | 0.08 | 0.06 | 0.04 | 0.04 | 1.00 | | | | | | | |
| NO-B | 0.04 | 0.04 | 0.02 | 0.03 | 0.72 | 1.00 | | | | | | |
| CO-1h | 0.17 | 0.15 | 0.17 | 0.16 | 0.06 | 0.04 | 1.00 | | | | | |
| CO-8h | 0.18 | 0.17 | 0.19 | 0.19 | 0.07 | 0.08 | 0.76 | 1.00 | | | | |
| FA-K | 0.00 | 0.00 | 0.00 | 0.01 | 0.00 | 0.00 | 0.03 | 0.04 | 1.00 | | | |
| FA-B | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.52 | 1.00 | | |
| АА-К | 0.00 | 0.00 | 0.02 | 0.03 | 0.00 | 0.00 | 0.03 | 0.06 | 0.12 | 0.05 | 1.00 | |
| AA-B | 0.00 | 0.00 | 0.02 | 0.03 | 0.00 | 0.00 | 0.02 | 0.04 | 0.10 | 0.13 | 0.76 | 1.00 |

 $^{^{}a}$ Indoor NO₂ and NO concentrations adjusted by subtracting 40% and 100%, respectively, of the simultaneous outdoor concentration. Indoor NO_x concentrations adjusted by summing the adjusted NO₂ and NO.

Table S5. Sample characteristics and median pollutant concentrations (ppb; except CO) in homes grouped by the type of gas appliance(s) in the living space. Symbols indicate statistical discernibility using Kruskal-Wallis test of likelihood that other groups' distributions are drawn from the same distribution as the first group: **p<0.01; * $0.01 \le p < 0.05$; $^{\circ}0.05 \le p \le 0.15$. Last column indicates discernible differences between the last two groups; only shows $p \le 0.15$.

| Parameter | No gas appliances (Ref. group) | Only vented gas appliances | Only gas cooking | Gas cooking + vented gas appliances | Compare last two groups |
|--|--------------------------------------|----------------------------------|------------------|-------------------------------------|-------------------------|
| Homes (N) | 38 | 28 | 144 | 142 | |
| Mean floor area (m ²) a | 115 | 159 | 148 | 111 | |
| Median floor area (m ²) ^a | 105 | 163 | 128 | 105 | |
| Measured concentrations b | | | | | |
| Highest 1-h CO (ppm) | 1.7 | 1.5 | 4.8** | 4.9** | |
| Bedroom NO _X | 20 | 23 | 54** | 69** | p=0.03 |
| Kitchen NO _X | 18 | 21 | 60** | 75** | p=0.09 |
| Bedroom NO ₂ | 6.7 | 6.0 | 14** | 16** | p=0.01 |
| Kitchen NO ₂ | 6.5 | 7.6 | 18** | 22** | p=0.05 |
| Bedroom formaldehyde | 13 | 18^ | 16 | 15 | |
| Kitchen formaldehyde | 14 | 16 | 16^ | 16 | |
| Bedroom acetaldehyde | 7.9 | 8.1 | 7.7 | 7.4 | |
| Kitchen acetaldehyde | 8.0 | 8.3 | 7.3 | 7.3 | |
| Indoor-attributed b,c | | | | | |
| Bedroom NO _X | -0.8 | 3.2 | 31** | 42** | p=0.02 |
| Kitchen NO _X | 3.9 | 5.6 | 38** | 53** | p=0.05 |
| Bedroom NO ₂ | 1.7 | 2.6 | 7.9** | 9.6** | p=0.02 |
| Kitchen NO ₂ | 1.4 | 3.2 | 12.3** | 16.9** | p=0.03 |
| Scaled to 130 m ² home ^a | | | | | |
| Highest 1-h CO (ppm) | 1.2 | 1.3 | 4.4** | 3.8** | p=0.07 |
| Indoor-attributed & scaled ^{a,b} | | | | | |
| Bedroom NO _X | -1.2 | 2.1 | 32** | 35** | |
| Kitchen NO _X | 3.2 | 4.8^ | 39** | 42** | |
| Bedroom NO ₂ | 1.1 | 2.7^ | 8.0** | 7.7** | |
| Kitchen NO ₂ | 1.5 | 3.0^ | 12.2** | 17.0** | |

^a Floor area assigned to each home for purpose of this calculation was the midpoint of the size bin selected by participant during telephone interview. Size bins were 46-70, 70-93, 93-116, 116-139, 139-186, 186-232, 232-279, >279 m². b Estimated by subtracting the assigned outdoor NO and 0.4x the outdoor NO₂ from corresponding kitchen and

bedroom measured values, then calculating indoor-attributed NO_X as NO+NO₂.

Table S6. Sample characteristics and median pollutant concentrations (ppb; except CO in ppm) in homes grouped by presence of pilot light(s) in the living space. Symbols indicate statistical discernibility using Kruskal-Wallis test of likelihood that other groups' distributions are drawn from the same distribution as the second group (homes with gas cooktop, no pilots): **p<0.01; *0.01 \le p<0.05; ^0.05 \le p \le 0.15. Last column indicates discernible differences between 3rd and 5th groups; only shows p \le 0.15.

| Parameter | Electric cooktop, no pilots | Gas cooktop, no pilots (Ref. group) | Gas cooktop w/pilot, no furnace pilot | Gas cooktop w/o pilot, furnace pilot | Gas cooktop w/pilot, furnace pilot | Compare 3 rd & 5 th groups |
|--|-----------------------------|---|---------------------------------------|--|--|--|
| Homes (N) | 60 | 182 | 29 | 48 | 27 | |
| Mean floor area (m ²) a | 138 | 165 | 115 | 86 | 79 | |
| Median floor area (m ²) ^a | 128 | 128 | 81 | 81 | 81 | |
| Measured concentrations | | | | | | |
| Highest 1-h CO (ppm) | 1.6** | 4.1 | 7.0* | 6.3^ | 4.8^ | |
| Bedroom NO _X | 20** | 51 | 78** | 70* | 108** | p=0.07 |
| Kitchen NO _X | 19** | 55 | 119** | 74* | 138** | |
| Bedroom NO ₂ | 6.5** | 12 | 26** | 18** | 32** | p=0.03 |
| Kitchen NO ₂ | 6.0** | 15.4 | 41** | 22** | 58** | p=0.14 |
| Bedroom formaldehyde | 14 | 16 | 17 | 12^ | 11^ | |
| Kitchen formaldehyde | 15 | 16 | 17 | 12* | 13 | |
| Bedroom acetaldehyde | 8.0 | 7.6 | 8.1 | 7.4 | 6.2 | |
| Kitchen acetaldehyde | 7.9 | 7.4 | 8.9 | 7.2 | 6.2 | |
| Indoor-attributed b | | | | | | |
| Bedroom NO _X | -0.1** | 26 | 62** | 41** | 109** | p=0.02 |
| Kitchen NO _X | 5.2** | 31 | 102** | 51** | 116** | p=0.09 |
| Bedroom NO ₂ | 1.9** | 6.5 | 16** | 9.9** | 28** | p<0.01 |
| Kitchen NO ₂ | 1.8** | 10.3 | 33** | 16* | 52** | p=0.03 |
| Scaled to 130 m ² home ^a | | | | | | |
| Highest 1-h CO (ppm) | 1.3** | 4.3 | 4.7 | 3.7 | 3.9 | p=0.13 |
| Indoor-attributed & scaled ^{a,b} | | | | | | |
| Bedroom NO _X | -0.4** | 28 | 42** | 26 | 58** | |
| Kitchen NO _X | 3.8** | 36 | 66** | 29 | 62** | |
| Bedroom NO ₂ | 1.6** | 7.1 | 11** | 5.9 | 16** | p=0.03 |
| Kitchen NO ₂ | 1.9** | 11 | 24** | 8.6^ | 22** | |

^a Floor area assigned to each home for purpose of this calculation was the midpoint of the size range that the participant selected during a telephone interview. Size ranges were 46-70, 70-93, 93-116, 116-139, 139-186, 186-232, 232-279, >279 m².

 $^{^{\}rm b}$ Estimated by subtracting the assigned outdoor NO and 0.4x the outdoor NO $_{\rm 2}$ from corresponding kitchen and bedroom measured values, then calculating indoor-attributed NO $_{\rm X}$ as NO+NO $_{\rm 2}$.

Table S7. Sample characteristics and median pollutant concentrations in homes grouped by cooking fuel type and amount of cooking during the week of sampling. Symbols indicate statistical discernibility using Kruskal-Wallis test of likelihood that other groups' distributions are drawn from the same distribution as the fourth group: **p<0.01; * $0.01 \le p<0.05$; $0.05 \le p\le0.15$.

| Parameter | Elec. CT, | Elec. CT, | Elec. CT, | Gas CT, | Gas CT, | Gas CT, |
|--|-----------|-----------|-----------|--------------|---------|---------|
| | <4h/wk | 4-8h/wk | >8h/wk | <4h/wk | 4-8h/wk | >8h/wk |
| | | | | (Ref. Group) | | |
| Homes (N) | 17 | 19 | 28 | 93 | 94 | 99 |
| Mean floor area (m ²) ^a | 105 | 138 | 164 | 120 | 130 | 141 |
| Median floor area (m ²) ^a | 81 | 114 | 163 | 105 | 105 | 128 |
| Measured concentrations | | | | | | |
| Highest 1-h CO (ppm) | 1.6** | 2.0* | 1.5** | 3.9 | 5.6** | 5.0* |
| Bedroom NO _X | 17** | 26* | 19** | 54 | 67 | 63* |
| Kitchen NO _X | 14** | 24** | 18** | 61 | 71^ | 74** |
| Bedroom NO ₂ | 6.6** | 7.3** | 5.2** | 13 | 14 | 17^ |
| Kitchen NO ₂ | 5.5** | 9.3** | 6.0** | 18 | 19 | 24^ |
| Bedroom formaldehyde | 19 | 15 | 13 | 14 | 16 | 15 |
| Kitchen formaldehyde | 16 | 15 | 14 | 14 | 16 | 17^ |
| Bedroom acetaldehyde | 7.8 | 9.2 | 7.6 | 7.5 | 7.9 | 7.4 |
| Kitchen acetaldehyde | 7.2 | 9.4^ | 7.4 | 7.1 | 7.4 | 8.5 |
| Indoor-attributed b | | | | | | |
| Bedroom NO _X | -1.6** | -0.9** | 0.6** | 24 | 46** | 47** |
| Kitchen NO _X | 4.5** | 5.2** | 3.7** | 28 | 52** | 59** |
| Bedroom NO ₂ | 2.7** | 1.9** | 1.7** | 7.2 | 8.8 | 11** |
| Kitchen NO ₂ | 1.3** | 3.2** | 2.1** | 12 | 13 | 17* |
| Scaled to 130 m ² home ^a | | | | | | |
| Highest 1-h CO (ppm) | 0.9** | 1.5* | 2.0* | 3.3 | 4.4** | 4.8** |
| Indoor-attributed & scaled a,b | | | | | | |
| Bedroom NO _X | -1.3** | -1.2** | 0.5** | 17 | 39** | 45** |
| Kitchen NO _X | 3.1** | 4.5** | 3.4** | 26 | 42** | 56** |
| Bedroom NO ₂ | 0.7** | 2.2** | 1.6** | 5.7 | 7.8^ | 10** |
| Kitchen NO ₂ | 0.8** | 2.6** | 2.3** | 9.8 | 13^ | 17** |

^a Floor area assigned to each home for purpose of this calculation was the midpoint of the size range that the participant selected during a telephone interview. Size ranges were 46-70, 70-93, 93-116, 116-139, 139-186, 186-232, 232-279, >279 m².

 $^{^{}b}$ Estimated by subtracting the assigned outdoor NO and 0.4x the outdoor NO $_{2}$ from corresponding kitchen and bedroom measured values, then calculating indoor-attributed NO $_{X}$ as NO+NO $_{2}$.

Table S8. Median pollutant concentrations in homes that cooked with gas for more than 4-h in during the monitoring period, grouped by the self-reported frequency of kitchen exhaust fan use. Results for homes with electric appliances included for comparison. Symbols indicate statistical discernibility using Kruskal-Wallis test of likelihood that other groups' distributions are drawn from the same distribution as the last group: **p<0.01; $*0.01 \le p < 0.05$; $^0.0.05 \le p \le 0.15$.

| Parameter | No gas cooking | Fan used most or all of the time | Fan used about half of the time | Fan used infrequently | Fan used rarely or never (Ref. group) |
|--|-------------------|----------------------------------|---------------------------------|-----------------------|---|
| Homes (N) | 64 | 46 | 31 | 61 | 55 |
| Mean floor area (m ²) a | 139 | 146 | 137 | 154 | 107 |
| Median floor area (m ²) ^a | 128 | 163 | 128 | 128 | 81 |
| Measured concentrations | | | | | |
| Highest 1-h CO (ppm) | 1.6** | 3.3** | 4.4* | 5.9^ | 7.6 |
| Bedroom NO _X | 20** | 57 | 54* | 62* | 93 |
| Kitchen NO _X | 19** | 57** | 64* | 70** | 105 |
| Bedroom NO ₂ | 6.3** | 14** | 16* | 14** | 24 |
| Kitchen NO ₂ | 6.6** | 16** | 22** | 20** | 34 |
| Bedroom formaldehyde | 15 | 18 | 16 | 15 | 15 |
| Kitchen formaldehyde | 15 | 17 | 17 | 17 | 14 |
| Bedroom acetaldehyde | 8.1 | 9.9^ | 8.4 | 7.1 | 6.7 |
| Kitchen acetaldehyde | 8.1^ | 9.4 | 8.6 | 7.2 | 6.6 |
| Indoor-attributed b | | | | | |
| Bedroom NO _X | -0.1** | 43** | 30** | 39** | 70 |
| Kitchen NO _X | 4.7** | 47** | 40** | 45** | 90 |
| Bedroom NO ₂ | 1.9** | 8.4** | 8.3** | 8.8** | 17.3 |
| Kitchen NO ₂ | 2.1** | 12** | 13** | 13** | 26 |
| Scaled to 130 m ² home ^a | | | | | |
| Highest 1-h CO (ppm) | 1.3** | 3.3^ | 4.1 | 5.2 | 4.7 |
| Indoor-attributed & scaled a,b | | | | | |
| Bedroom NO _X | -0.7** | 46 | 31* | 38^ | 50 |
| Kitchen NO _X | 3.5** | 44** | 39* | 42* | 64 |
| Bedroom NO ₂ | 1.5** | 8.5* | 7.9^ | 9.5* | 11 |
| Kitchen NO ₂ | 1.7** | 11** | 13* | 15** | 19 |

^a Floor area assigned to each home for purpose of this calculation was the midpoint of the size range that the participant selected during a telephone interview. Size ranges were 46-70, 70-93, 93-116, 116-139, 139-186, 186-232, 232-279, >279 m².

 $^{^{}b}$ Estimated by subtracting the assigned outdoor NO and 0.4x the outdoor NO₂ from corresponding kitchen and bedroom measured values, then calculating indoor-attributed NO_X as NO+NO₂.

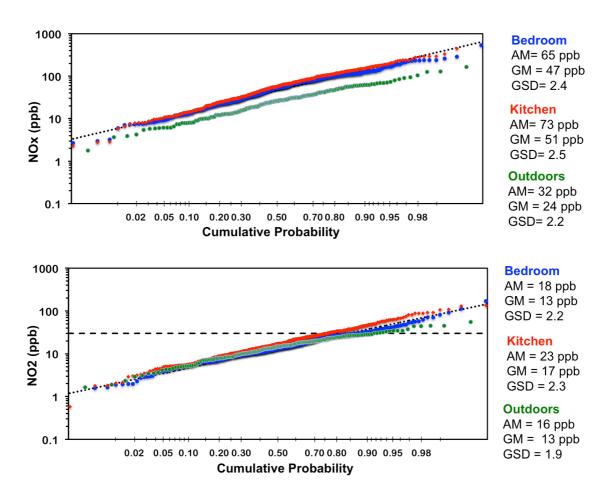


Figure S1. Measured concentrations of NO_x and NO₂ inside and outside of study home.

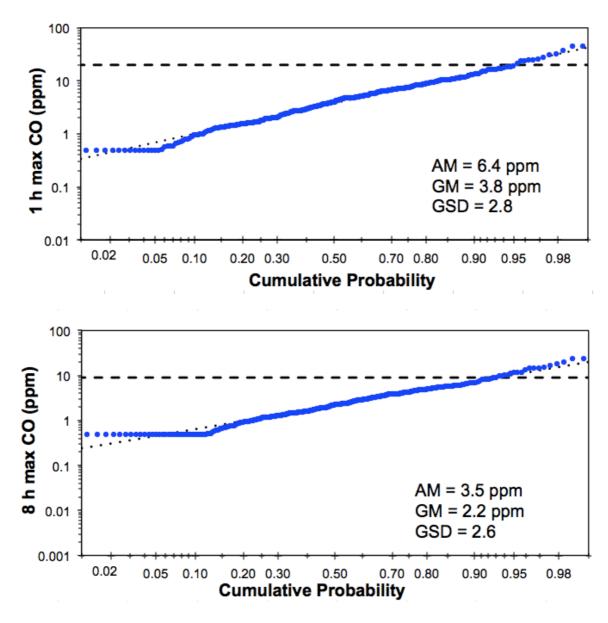


Figure S2. Highest 1h and 8h CO measured in kitchens of study homes.

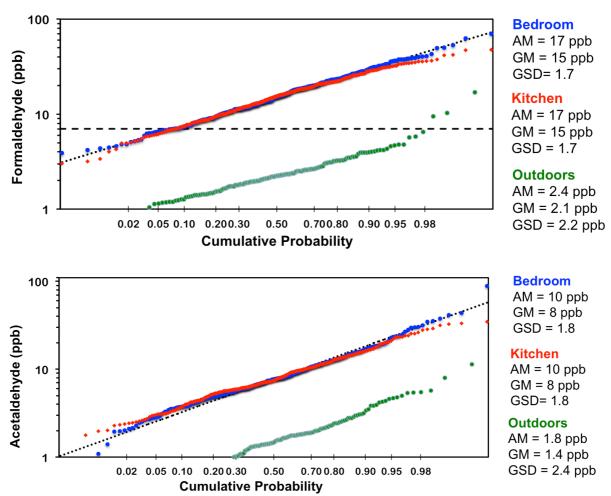


Figure S3. Measured concentrations of formaldehyde and acetaldehyde inside and outside of study homes. As a group, outdoor acetaldehyde concentrations were indistinguishable from field blanks.

Comparison of Pollutant Concentrations Measured in the Healthy Homes Study of 2011-2013 to Other Studies of California Homes

Comparisons are presented in text and NO₂ statistics are summarized in Table S9.

Both outdoor and indoor NO₂ concentrations measured in the Healthy Homes study of 2011-2013 were lower than those reported for California studies in the 1980s and early 1990s. (Wilson et al., 1986) reported results of one-week passive measurements of NO₂ in kitchens, bedrooms, and outdoors at over 600 homes in Southern California in March and July of 1984 and January of 1985. (Spengler et al., 1994) reported two-day measurements for a population representative sample of 482 households in a nitrogen dioxide exposure study in Los Angeles in 1987-88. (Wilson et al., 1993) reported NO₂ measured by passive samplers deployed over 48 h periods at homes throughout California's investor-owned utility service areas in 1991-92. Relevant comparisons are provided below by noting statistics from each study for homes grouped by appliances found to impact NO₂ concentrations in these studies.

In January 1985, Wilson et al. (1986) reported (in their Table 6-8) that homes with all electric appliances (n=13) had median outdoor, bedroom, and kitchen NO₂ levels of 49, 19, and 23 ppb. Homes with electric ranges and gas forced air furnaces (n=68, their Table 6-9) had median outdoor, bedroom, and kitchen NO₂ of 51, 24, and 29 ppb. In the winter of 1987-88, homes with electric cooking (n=46) had median outdoor and bedroom NO₂ concentrations of roughly 40 and 20 ppb (Figure 3 of Spengler et al., 1994). All-electric homes in the Healthy Homes Study of 2011-13 had median NO₂ of 8.6 ppb outdoors, 6.7 ppb in bedrooms, and 6.5 ppb in kitchens.

In January 1985, homes with forced air furnaces and gas ranges without pilot burners (n=38) had median outdoor, bedroom, and kitchen NO₂ of 54, 30, and 41 ppb (Table 6-9 of Wilson et al. (1986)). In winter 1987-88, homes with gas cooktops without pilot burners (n=34) had median NO₂ levels of 41 ppb outdoors and 22 ppb in bedrooms (based on Figure 3 of Spengler et al. (1994)). In 2011-13, homes with gas cooking without pilots had median outdoor, bedroom, and kitchen NO₂ of 14, 14, and 17 ppb.

In January 1985, homes with gas cooking burners with pilots (n=98) had median outdoor, bedroom and kitchen NO₂ of 60, 43, and 63 ppb (Table 6-9 of Wilson et al. (1986)). In winter 1987-88, homes with gas cooking burners with pilots (n=93) had median NO₂ of 45 ppb outdoors and 32 ppb in bedrooms (Figure 3 of Spengler et al. (1994)). In 2011-13, homes with gas cooking appliances with pilots had median outdoor, bedroom and kitchen NO₂ of 13, 29, and 46 ppb.

Wilson et al. (1993) reported that almost all of their study home had gas appliances but they did not report NO₂ resolved by home appliance characteristics; they reported median NO₂ of 25 ppb outside and 16 ppb in bedrooms. The Logue et al. (2014) simulation study of Southern California homes that cook with gas at least once per week reported a median time-integrated household NO₂ concentrations of 16 ppb (which is similar to 2011-13 measurements) and median outdoor NO₂ of 24 ppb (which is higher than the 2011-13 data).

Estimates of the indoor-attributed NO₂ in homes using natural gas cooking burners in 2011-2013 are generally similar to estimates from the data reported by Wilson et al. (1986). Using an infiltration factor of 0.46 based on NO₂ measured outside and in bedrooms of homes with

electric cooking (Figure 6-21 of Wilson et al. (1986)), the estimated median indoor-attributed NO₂ for all homes with gas cooking in January 1985 is 48–0.46*64=18 ppb. The comparable value for kitchens is 69–0.46*64 = 39 ppb. Focusing only on homes with natural gas forced air furnaces in the January 1985 sample, we estimate median indoor-attributed NO₂ levels of 5.3 and 17 ppb in bedrooms and kitchens of (n=38) homes with gas cooking without a pilot. In 2011-13, median indoor-attributed NO₂ levels were 7.4 and 12 ppb in bedrooms and kitchens of homes with gas cooking without pilots. From the 1985 data, we estimated median indoor-attributed NO₂ levels of 15 and 36 ppb for bedrooms and kitchens of (n=98) homes with piloted gas cooking. In 2011-13, homes with piloted gas cooking appliances had median bedroom and kitchen NO2 levels of 19 and 36 ppb. (These values are summarized in Table S-9).

Aldehyde concentrations measured during winter sampling in this study are similar to those reported for sampling throughout the year in 105 homes in Los Angeles CA (Weisel et al., 2005); that study reported median indoor and outdoor concentrations of 15.4 and 5.3 ppb for formaldehyde, and 11.5 and 2.9 ppb for acetaldehyde. Median indoor concentrations of formaldehyde and acetaldehyde during winter sampling in 17 homes were higher (20.6 and 12.5 ppb, respectively), although outdoor concentrations were lower (2.9 and 2.3 ppb). Formaldehyde levels measured in 2011-13 were substantially lower than those reported for a mail-out study of 51 San Francisco Bay Area homes in January 1984 (Sexton et al., 1986). That study reported GM (GSD) values of 36 ppb (1.64) and 32 ppb (1.66) for kitchens and bedrooms, respectively.

Table S9. Comparisons between median NO2 concentrations (ppb) measured in Healthy Homes Study of 2011-13 and prior studies conducted in California.

| Home Group | Location | Data from 1985 | Data from 1987-88 | Data from 2011-13 |
|-------------------------------|--------------------------|----------------------|----------------------|-------------------|
| All electric / electric range | | N=13 / 68 a | N=46 | N=38 |
| | Outside | 49 / 51 ^a | 40 | 8.6 |
| | Bedroom | 19 / 24 ^a | 20 | 6.7 |
| | Kitchen | 23 / 23 ^a | n/a | 6.5 |
| Gas range no pilot | | N=38 | N=34 | N=223 |
| | Outside | 54 | 41 | 14 |
| | Bedroom | 30 | 22 | 14 |
| | Kitchen | 41 | n/a | 17 |
| | Bedroom, indoor source b | 5.3 | n/a | 7.4 |
| | Kitchen, indoor source b | 17 | n/a | 12 |
| Gas range with pilot | | N=98 | N=93 | N=56 |
| | Outside | 60 | 45 | 13 |
| | Bedroom | 43 | 32 | 29 |
| | Kitchen | 63 | n/a | 49 |
| | Bedroom, indoor source b | 15 | n/a | 19 |
| | Kitchen, indoor source b | 36 | n/a | 39 |

^a First group is for homes with all electric appliances / Second group is Electric range and gas forced air furnace. ^b Indoor-attributed values subtract the estimated contribution of outdoor NO2 to indoor concentrations. N values for indoor-attributed in 2011-13 study are n=229 for gas ranges without pilots and n=45 for gas ranges with pilot.

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